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Interference of direct and resonant channels in electron-impact ionization of C^{3+} ions: Unified *R*-matrix calculation and experiment

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Calculated total cross sections for electron-impact ionization of C^{3+} using a unified theoretical approach are compared with high-resolution high-precision measurements clearly demonstrating interference of resonant multielectron reaction channels with direct ionization and inner-shell excitation. In particular, the so-called READI process, i.e., *K*-shell excitation with simultaneous capture of the incident electron (resonant-excitation) and a subsequent three-electron interaction resulting in simultaneous ejection of two electrons (auto-double-ionization) has been studied in unprecedented detail. The calculation is in very good agreement with the experimental cross section.

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Electron-ion collisions are important in all plasma environments. Hence, besides the fundamental interest in atomic interactions and structures, the fields of astrophysics and applied plasma research are pushing experimental and theoretical investigations of electron-impact excitation, ionization, and recombination. Vast applied data needs for cross sections, rate coefficients, and other microscopic quantities exist that can never be met by experiment alone. Theory has to fill the gap, but the calculations have to be checked by experiment in order to prove their reliability. Resonance phenomena in electron-ion collisions and the principle possibility of interference between direct and indirect reaction channels provide an ideal testing ground for advanced theoretical models.

In this paper we report on detailed theoretical and experimental studies of electron-impact ionization of C^{3+} ions. Using a recently developed unified theoretical approach and a new electron-ion crossed-beams setup, a unique view can be taken at the interplay of complex resonant and nonresonant excitation channels with direct ionization (DI).

The ionization of lithiumlike ions is dominated by direct removal, DI, of the outer $2s$ electron, whereas DI of the *K* shell contributes little to the total cross section. Although conceptually simple, DI still provides unresolved problems. While theory and experiment are in very good agreement for (direct) ionization of hydrogenlike ions [1,2], recent massive theoretical efforts (see, e.g., [3–5]) in calculating DI of Li-like and Na-like ions have revealed confusing discrepancies with existing experiments. In most recent publications this dilemma was discussed also for the case of C^{3+} [6,7], where theory is about 20–30 % above the measurements [8–10].

An additional reaction channel beside DI is the excitation of a $1s$ electron to a bound nl state producing a doubly excited configuration $1s2snl$ that decays by autoionization. It has been established previously that this process, termed excitation autoionization (EA), contributes about 5 to 10 % of the cross section for C^{3+} above the *K*-shell excitation threshold [8,10,11]. These findings were in agreement with theoretical calculations (e.g., those of Younger [12]) of *K*-shell excitation cross sections. The EA process is accompanied by resonant-excitation double-autoionization (REDA) and resonant-excitation auto-double-ionization (READI). Both processes include resonant capture of the incident electron and subsequent emission of two electrons: sequentially in the REDA process and simultaneously in the READI process.

First experimental evidence for REDA and READI processes was found by Müller *et al.* [10,13]. Linkemann *et al.* [14] experimentally verified that REDA can even dominate ionization. Theoretical work on resonance contributions to the ionization cross section is sparse. REDA calculations for C^{3+} have been performed by Tayal and Henry [15] using a close-coupling technique, and by Reed and Chen [16] using the isolated resonances and independent processes approximation in a perturbative approach. The calculated results agree quite well with the experiment of Müller *et al.* [10].

In contrast to that, the much more sophisticated READI process has not yet been treated in a satisfactory manner. A prediction had been worked out by Pindzola and Griffin [17], who used first- and second-order perturbative theory to provide a rough estimate of the strength of a single READI resonance arising from the $1s2s^22p\ ^3P$ intermediate term. The main difficulty with such calculations is obtaining accurate double-Auger rates. Moreover, the possible interference of different reaction channels was neglected. Neither of the above methods gave a full explanation of all the resonant features observed in the experiment. In particular, due to the inherent deficiencies, none of the available theoretical models was able to explore the READI resonances occurring below the EA threshold. Recently, Berrington *et al.* [18] proposed a unified theoretical treatment to calculate REDA and READI contributions to electron-impact ionization of Be^{+}. However, this method did not completely account for resonance features, and it could not yet be tested against experiment.

In a combined theory-and-experiment effort, we have performed calculations and measurements for electron-impact ionization of C^{3+} ions, with special emphasis on the energy range 220–360 eV where resonances associated with the excitation of one *K*-shell electron can contribute. Our results show cases where the independent process approximation breaks down. Interference of REDA and EA is clearly seen and interference between READI and DI is unambiguously observed. Moreover, the experiment, in which isotopically clean $^{13}C^{3+}$ has been used, avoids a weakness of several previous measurements with C^{3+} ions: the problem of non-separable beam components $^{12}C^{3+}$ and $^{16}O^{4+}$. The measurements yield cross sections for single ionization that are about 30% above previous results. For an additional test of the validity of the experimental procedures, absolute ionization cross sections of hydrogenlike He^+ have been determined both before and after the C^{3+} measurements. The results obtained for He^+ are in perfect agreement with distorted-wave-exchange (DWE) calculations [12] and with the previous measurements of Peart *et al.* [19]. The present results for C^{3+} with their total uncertainty of about 12% are slightly above the latest and most advanced theoretical calculations for DI of C^{3+} [6,7], including the present theoretical approach.

The experiments were carried out at a new crossed-beams setup for measurements of total single and multiple ionization of atoms and ions by electron impact. The measurement of total cross sections follows the technique developed by Müller *et al.* [20]. An ion beam produced by a permanent-magnet ECR (electron-cyclotron-resonance) ion source is crossed with an intense electron beam that is mechanically moved through the ion beam for absolute cross-section measurements and that is is set to optimum beam overlap for fast energy scans (see [10,13]). The potential of the technology has been well documented previously.

A complete theoretical description of both direct and indirect ionization processes demands a unified total wave function. The *R*-matrix method [21] uses a close-coupling approach, in which the total $(N+1)$-electron wave function is expanded in terms of an *N*-electron target basis, which contains the required initial and final states and other states strongly coupled to these. However, such an expansion over target bound states is not complete if the probability of ionization is significant: in which case there should also be included an integral over the continuum. A convenient way of representing this integral while retaining the simplicity of the close-coupling expansion is to use a pseudostate discretization to approximate the quadrature [22]. Such an approach has been shown to give good convergence over a wide range of energies (see, e.g., [6]), but can involve large sets of pseudostates; at present it would be computationally difficult to also include inner-shell autoionizing states in these expansions. However, Berrington *et al.* [18] suggested that at a given (high) energy *E*, the open channel part of the integral that is continuous in the energy range below *E* can be formally approximated by a mean value in the range. They therefore examined the possibility of introducing energetically allowed pseudostates $\overline{nl}$ for each target angular momentum l to approximate the direct cross section into the continuum, and found for Be^+ and Li^+ that suitable pseudostates could be constructed from orbitals optimized on the inner-shell states such that the total cross section becomes $\sigma_{\text{ionization}}(E)\approx\Sigma_{\overline{nl}}\sigma_{\overline{nl}}+\sigma_{\text{autoionization}}$. This approximation is clearly energy dependent, but providing we are in a region where the direct ionization cross section varies only slowly with energy, then the approximation may be good over a reasonable range of energies. Berrington *et al.*, however, found that the ionization cross section was likely to be overestimated in this approach unless a further pseudostate, optimized on the dipole polarizability of the ground state, was included in the bound-state spectrum to allow for loss of flux into the infinity of dipole-coupled bound states.

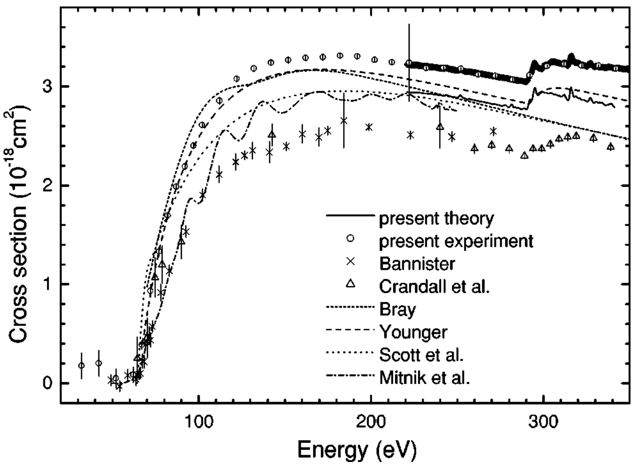

FIG. 1. Ionization cross sections for C^{3+}. Besides the present results experimental data of Bannister [9] and Crandall *et al.* [8] as well as theoretical results of Bray [4], Mitnik *et al.* [6], Scott *et al.* [7] and Younger [12] are displayed. The long error bars on some of the experimental data show typical total experimental uncertainties; short error bars are statistical.

In the present calculation, we adopt the same procedure; however, different from the work of Berrington *et al.* [18], a real $3p$ orbital and a $\overline{5p}$ polarized orbital are included. The target states consist of five physical states of C^{3+}, five pseudostates and 16 autoionizing states. All target states were represented by configuration interaction (CI) wave functions. Eleven orbitals were used: the $1s$, $2s$, $2p$, $3s$, $3p$, and $3d$ orbitals were taken from the table given by Weiss [23]; the $\overline{4s}$, $\overline{4p}$, $\overline{4d}$, and $\overline{4f}$ orbitals were optimized on the $1s2s^2$, $1s2p^2$, and $1s2s2p$ inner-shell excited states, using the CIV3 package of Hibbert [24]; and the $\overline{5p}$ orbital was optimized on the $1s^22s$ ground-state dipole polarizability. The target orbitals require an *R*-matrix radius of 10.0 a.u. Twenty-four continuum basis functions were used per angular momentum. Partial waves up to total angular momentum $L=20$ were needed to obtain converged results for the ionization cross sections. The internal region *R*-matrix package RMATRX II [25] and the external asymptotic program STGF [26] were employed. The resonance positions and widths were determined with a method introduced by Quigley and Berrington [27]. Radiation damping was neglected, which should be a good approximation for C^{3+} ions.

A comparison of our theoretical and experimental cross-section results is shown in Fig. 1, together with the DI calculations of Bray [4], Mitnik *et al.* [6], and Scott *et al.* [7], and with Younger's DWE calculation [12], which includes DI of the $2s$ and $1s$ subshells as well as nonresonant EA channels. Also shown are previous absolute experiments

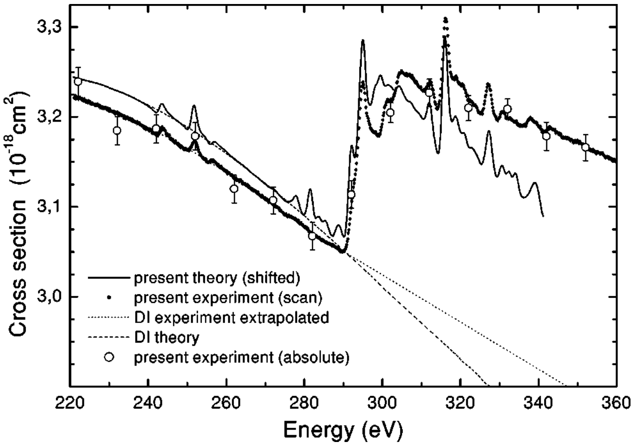

FIG. 2. Detail of Fig. 1. The present theory data are convoluted with a 1.8-eV FWHM Gaussian and shifted up by 3×10^{-19} cm^2; present scan data are shown with statistical error bars. The dashed and dotted lines are extrapolated DI contributions to the present data.

[8,9]. Below 290 eV the cross section is almost entirely determined by DI. In that energy range the present experimental cross sections are slightly above the DI-only calculations and the present unified theory (spanning only a limited energy range due to the complexity of the unified approach); however, the measurements include all theoretical results within their total uncertainty. Above 290 eV both the experimental data and the unified theory clearly show the well-known onset of EA contributions.

A closer look is taken at the EA threshold region in Fig. 2. Here, the experimental energy scan data are displayed and compared with the unified calculation. The experimental energy axis was calibrated against the theoretical energy of the $1s2s^22p\ ^3P$ state at 243.05 eV; i.e., the experimental energy scale was shifted down by 1.05 eV. The theoretical cross sections have been convoluted with a 1.8-eV full width at half maximum (FWHM) Gaussian that accounts for the experimental energy spread. For easier comparison, the present theoretical cross sections have been shifted up in this plot by 0.3×10^{-18} cm^2. Theory and experiment agree in most of the cross-section details. However, there are differences in the energy dependence of DI. DI energy dependences are inferred from both sets of data, resulting in the smooth curves displayed in Fig. 2. Apparently, the theoretical DI curve is slightly steeper than the experimental one. Similar differences occur between the theoretical data sets displayed in Fig. 1. On the experimental side there is a limited reproducibility of single absolute cross-section measurements within 2%, while for the observation of fine structures in the cross-section only statistics is setting the limits. Since the scan results have to be normalized to the absolute measurements, there remains a systematic uncertainty of the overall scan-data energy dependence, while the point-to-point uncertainty of the measurements is less than 0.04% in the present case. Discrepancies also occur just below the EA threshold where the unified theory predicts READI resonances that are not observed with the predicted strengths in the experiment. Further theoretical work is needed to clarify this situation.

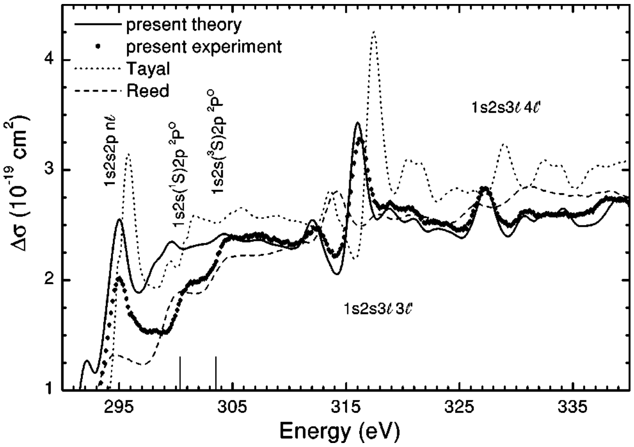

FIG. 3. Cross sections from which the DI ‘‘background’’ (from Fig. 2) has been subtracted. The region of dominant REDA resonances is shown. Previous calculations by Reed and Chen [16] and Tayal and Henry [15] are also displayed.

For more detailed comparisons of the fine step and peak features in the indirect cross-section contributions we subtracted the smooth DI ‘‘background’’ curves of Fig. 2 from the experimental and theoretical data sets, respectively. When the resulting ‘‘indirect’’ ionization cross sections $\Delta\sigma$ are displayed in the next two figures without any adjustment factors or shifts, one has to bear in mind the uncertainty of the subtraction procedure, which could also produce results $\Delta\sigma$ differing, e.g., by 20–30 % at 340 eV. Figure 3 shows $\Delta\sigma$ in the region where the dominant EA and REDA contributions occur. The present unified theory is in good agreement with the experiment.

At $\sim$314 eV there is a clear dip in the cross section seen in both our data sets. The shape of the resonances in this vicinity and the fact that the dip goes below the average nonresonant course of the cross section indicate the presence of destructive interference. This interpretation is strongly supported by the comparison of the present data with the independent-process approximation of Reed and Chen [16], which shows no dip anywhere around 314 eV. Clearly, by neglecting interference, a prominent feature in the experiment cannot be reproduced. We attribute the window at 314 eV to interference of the EA reaction path with the strong REDA resonance channel associated with $1s2s3l3l'$ configurations. This is supported by the *R*-matrix calculations of Tayal and Henry [15], who treated direct and indirect ionization as separable processes but allowed for interference between EA and REDA. Their result clearly shows a peak-and-dip feature at $\sim$316 eV that is almost identical to the present findings at $\sim$314 eV. Since they did not include DI, the destructive-interference dip in $\Delta\sigma$ can only be due to interacting EA and REDA channels.

What we consider the most important accomplishment of the present study is documented in Fig. 4, where the energy range of READI processes is investigated. The prominent features in $\Delta\sigma$ arise from $1s2s^22p\ ^3P$ and $1s2s2p^2\ ^3D$ resonances that contribute to ionization via simultaneous emission of two electrons. Experimentally, these features could only be made visible by reducing the relative uncer-

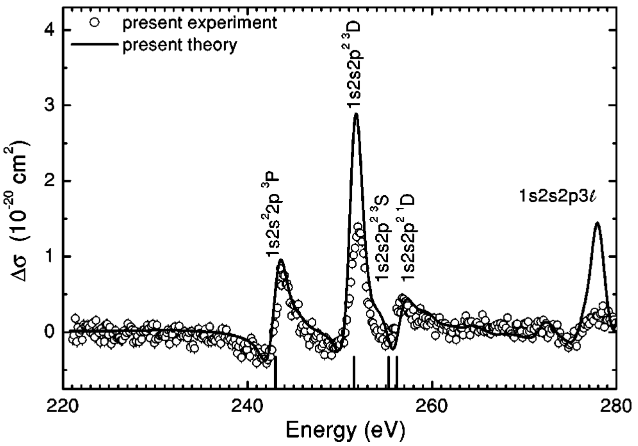

FIG. 4. Cross sections from which the DI ''background'' (from Fig. 2) has been subtracted. Theoretical energies of the dominant READI resonances are indicated.

tainties of the measured cross sections to the present level of 0.04 %. Theoretically, READI had not been treated in a satisfactory manner before the development of the present *R*-matrix techniques. In our results we note an asymmetry of the dominant READI resonances that is even accompanied by slight excursions of $\Delta\sigma$ to negative values, clearly indicating patterns of interference between READI and DI channels (the latter was subtracted from the total cross section; EA and REDA are energetically forbidden in the energy range of Fig. 4). The asymmetric interference profiles are seen in the unified calculations and in the experiment alike.

It is obvious that only unified theoretical approaches provide accurate tools to calculate details of ionization cross sections. Such details are the interference patterns observed in the experimental energy ranges selected in Figs. 3 and 4. Clearly the present investigation shows that READI interferes with DI and that REDA interferes with the EA channel.

In summary, for electron-impact ionization of C^{3+} ions we present results of a unified *R*-matrix approach, including all direct and indirect mechanisms as well as their possible interactions. Accurate calculations of the READI mechanism have been carried out and tested against detailed experiments. We have performed absolute cross-section measurements with $^{13}C^{3+}$ ions, avoiding previous problems arising from ion-beam contaminations with $^{16}O^{4+}$. Thereby we have obtained results that might explain previously observed discrepancies between existing experimental data and the most advanced theoretical approaches. Energy scan measurements with very much improved statistics were performed, which in conjunction with unified *R*-matrix calculations allowed us to study details of READI resonances in the electron-impact ionization of C^{3+} ions. The shape of the most pronounced resonances clearly shows the importance of the interference of the READI channel with direct ionization.

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